United States Environmental Protection Agency **Office of Research and Development**

National Exposure Research Laboratory
Research Abstract

Government Performance Results Act (GPRA) Goal 1
Annual Performance Measure #337

Significant Research Findings:

Evaluate Models-3/CMAQ for Particulate Matter using Episodic Data

Scientific Problem and Policy Issues

Ambient air concentrations of particulate matter (PM) continue to be a major concern for the Agency. High concentrations have been linked to detrimental health effects, acid precipitation, regional climate change and visibility degradation. Accordingly, the Clean Air Act and Amendments call for an assessment of the impacts of current and future regulations designed to protect human health and welfare. The most reliable tools for carrying out such assessments are air quality models such as Models-3 Community Multiscale Air Quality (CMAQ) model. In order to establish credibility and build confidence within the user and scientific community, CMAQ like all deterministic models needs to be evaluated through comparisons with observations. Accordingly, the purpose of this research is to thoroughly evaluate and characterize the performance of the CMAQ system and its emissions, meteorological and chemical/transport modeling components, with an emphasis on air concentrations of fine particles, which is central to the 2005-2008 SIP implementations.

Research Approach

Characterization of the model's performance involved two types of evaluation. The first type was an *operational evaluation*, which utilized data from routine, nationwide monitoring networks (e.g., the Speciated Trends Network (STN), the Clean Air Status Trends Network (CASTNet)) to determine if the PM concentrations of regulatory interest are adequately being simulated (*Is the model providing the right answer?*). These are fairly dense networks that collect data on a long-term continuous basis, but the data are typically daily or weekly average values. The second type was a *diagnostic evaluation*, which utilized intensive, regional field studies (e.g., the Nashville Southern Oxidant Study, Atlanta Supersite Experiment, EPA Supersite Program) that collect a broader range of chemical species on an hourly basis to examine the ability of the model to accurately simulate all of the physical and chemical processes that lead to the simulated concentration (*Is the model providing the right answer for right reason?*). In addition to direct evaluations, model sensitivity analyses were also conducted as part of this task to characterize model response to uncertainties.

Results and Impact

Two simulation periods (4 January - 19 February 2002 and 15 June - 16 July, 1999) were selected for evaluation of the CMAQ model performance for fine particulate matter. Observational data from four networks were used: the Clean Air Status and Trends Network (CASTNet), the Interagency Monitoring of Protected Visual Environments (IMPROVE) network, the Speciated Trends Network (STN),

and the Southeastern Aerosol Research and Characterisation (SEARCH) network. A series of evaluation tests were performed using the FY02 public release version and beta-versions of the eventual FY03 public release version of the Models-3/CMAQ. With the exception of nitrate, the FY02 CMAQ simulation results indicated generally good performance, especially for sulfate and ammonium where, depending on network and episode, correlations were generally quite good (e.g., correlation coefficients > 0.70). Nitrate, on the other hand, was greatly over-predicted by the model. The FY02 CMAQ's simulations of nitrate were deemed unacceptable, and the correction to this "nitrate problem" was set as a top priority for the FY03 public release of the model.

In order to identify the cause(s) for the nitrate overprediction, several diagnostic evaluations were performed using highly time resolved (hourly) data obtained at the Atlanta and Pittsburgh Supersites. These evaluations showed the FY02 CMAQ had very large over-predictions of nitric acid and/or total nitrate that peaked at night, suggesting an issue with the nighttime heterogeneous production of nitric acid on wetted aerosol particles. Reaction rates used in the FY02 CMAQ for this nighttime pathway for nitric acid production were two to three orders of magnitude higher than the probability estimates from the most recent literature. A test of these new literature values for the heterogeneous reaction probabilities showed a dramatic improvement in the predictions of CMAQ for nitric acid and aerosol nitrate, although CMAQ was still over-predicting nitric acid at night. Sensitivity studies showed that eliminating altogether the nighttime heterogeneous production of nitric acid in CMAQ brought its predictions in line with the nighttime levels of nitric acid at both special sites. The comparisons of these CMAQ sensitivity runs now showed a modest over-prediction of nitric acid occurred during the day, implying that the photochemical mechanisms for ozone production are also creating part of the nitrate over-prediction problem. The episodic evaluations after the modifications to the heterogeneous production of nitric acid in CMAQ indicate that the heterogeneous reaction probability is still too high; thus, the "nitrate problem" has been greatly ameliorated, but not eliminated. There are suggestions in the laboratory research community that a variety of factors exist that further inhibit these nighttime reactions, but none are published and available for use by the CMAQ developers at this time.

A serious deficiency in the CMAQ simulation was, to an acceptable degree, fixed with the help of these episodic evaluations and sensitivity tests. Changes to the nighttime heterogeneous reaction rate for production of nitric acid were incorporated into the FY03 public release version of CMAQ, and episodic comparisons against observational networks and special hourly sites indicated improvements to nitrate predications as a result of these changes. However, CMAQ still shows a moderate, systematic over-prediction of total nitrate and, hence, particulate nitrate, based on comparisons against the CASTNet data. It is important that continued improvements are pursued because CMAQ is expected to predict ammonia limitation more often than it should in certain areas of the country because of these total nitrate over-predictions. Thus, the FY03 and FY04 release versions of CMAQ could over-emphasize the nitrate replacement that can offset part of the reduction in sulfate that will accompany reductions of SO_2

emissions. As the state of science for nitrate aerosol predictions continues to improve, these potential impacts on regulatory decision making will be further tested against episodic data to better understand these interactions and impacts.

Research Collaboration and Research Products

Division staff conducted the model evaluation studies. Network data (e.g., CASTNet, STN, etc.) were obtained through public access web sites. Episodic data such as the Nashville SOS intensive were obtained directly from the Principal Investigators who maintain the data. Key, recent presentations and publications are provided below:

Dennis, R. L. "CMAQ winter predictions of nitrate: the importance of N_2O_5 reactions to HNO_3 production". Presented at NOAA's Aeronomy Laboratory, June, 2003.

Dennis, R. L. "Time resolved and in-depth evaluation of PM and PM precursors using CMAQ". Presented at EPA's PM Model Performance Workshop, Chapel Hill, NC. February 2004.

Arnold, J.R., R.L. Dennis, and G.S. Tonnesen. Diagnostic evaluation of numerical air quality models with specialized ambient observations: Testing the Community Multiscale Air Quality modeling system (CMAQ) at selected SOS 95 ground sites. *Atmospheric Environment* 37: 1185-1198 (2003).

Yu, S, Dennis, R. L., Roselle, S. J. Nenes, A., Walker, J., Eder, B. , Schere, K.and Swall, J. "An assessment of the ability of 3_D air quality models with current thermodynamics equilibrium models to predict aerosol nitrate." *Journal of Geophysical Research.* American Geophysical Union, Washington, DC, 110(D7):1-22, (2005).

Future Research

Future episodic model evaluation research will focus on both winter and summer months and will use data from the EPA Supersite Program (1) to examine the diurnal performance of CMAQ on several inorganic and primary species and (2) to further examine the implications of the nighttime over-prediction of nitric acid on the control strategy response of CMAQ to reductions in SO_2 emissions. In the latter work, indicators of the inorganic system state (whether it is ammonia limited or nitric acid limited) will be constructed from CMAQ predictions and compared against indicators derived independently from measurements, in order to assess the expected reliability of the control strategy predictions of CMAQ for inorganic species and to evaluate the indicators as diagnostic metrics.

Contacts for Additional Information

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